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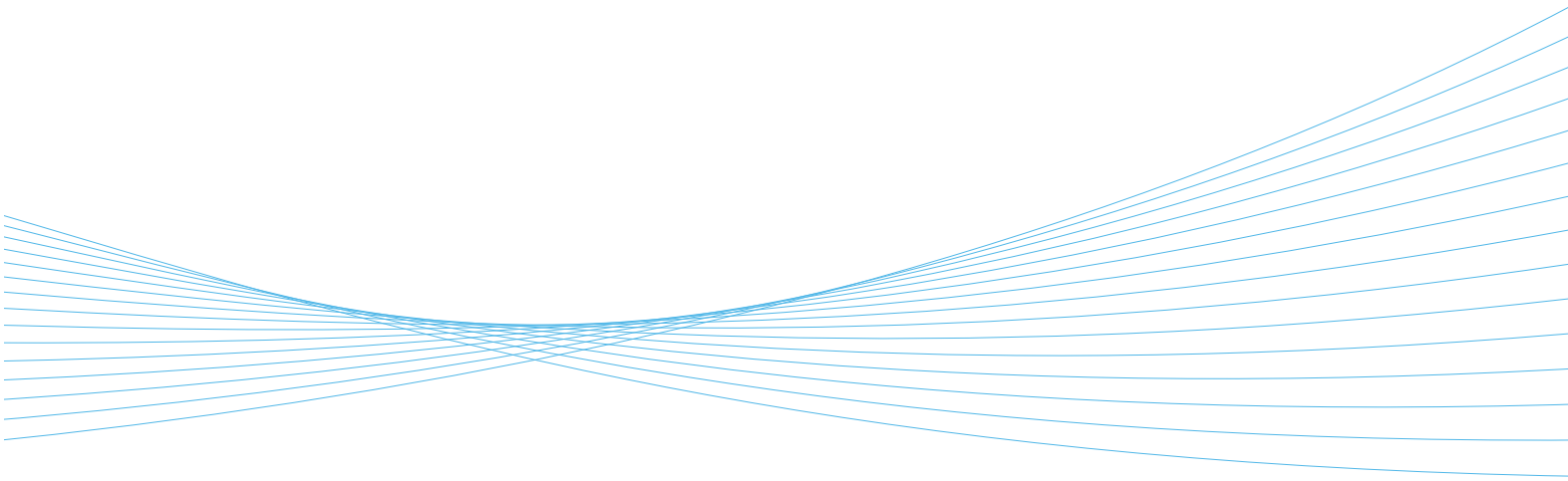
CLIMATIC EFFECT OF LIGHT-ABSORBING IMPURITIES ON SNOW: EXPERIMENTAL AND FIELD OBSERVATIONS

JONAS SVENSSON



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Climatic effect of light-absorbing impurities on snow: experimental and field observations

Abstract
Snow and ice are essential components of the Earth system, modulating the energy budget by reflecting sunlight back into the atmosphere, and through its importance in the hydrological cycle by being a reservoir for fresh water. Light-absorbing impurities (LAI), such as black carbon (BC) and mineral dust (MD), have a unique role in influencing the reflectance of the cryosphere. Deposition of the anthropogenic and natural LAI constituents onto these bright surfaces initiates powerful albedo feedbacks that will accelerate melt. This is important globally, but especially for regions such as the Arctic and the Himalaya.
In this thesis, observations from both ambient and laboratory experiments are presented. The overarching research goal has been to better understand the climatic effect of LAI on snow. More specifically, an emphasis has been placed on exploring the process-level interactions between LAI and snow, which will enable better comprehension of LAI affecting the cryosphere.
Key findings in this thesis involves the investigations on the horizontal variability of BC concentrations in the surface snow that indicate a larger variability on the order of meter scale at a pristine Arctic site compared to a polluted site nearby a major urban area. In outdoor experiments, where LAI were used to artificially dope natural snow surfaces, the snow albedo was observed to decrease following LAI deposition. The albedo decrease was on the same order as in situ measurements of LAI and albedo conducted elsewhere. As snow melted during the experiment, the snow density was observed to decrease with increasing LAI concentration, while this effect was not observed in non-melting snow. The water retention capacity in melting snow is likely to be decreased by the presence of LAI. Measurements examining the absorption of BC indicate that BC particles in the snow have less absorbing potential compared to BC particles generated in the laboratory. The LAI content of snow pit investigations from two glaciers in the Sunderdhunga valley, northern India, an area not previously examined for LAI, presented high BC and MD content, affecting the radiative balance of the glacier snow. At different points, MD may be greater than BC in absorbing light at the snow surface. A continued monitoring of LAI in the cryosphere, both on the detailed scale explored here, as well as on the larger modelling perspective is needed in order to understand the overall response of the cryosphere to climate change.

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CONTENTS

LIST OF PUBLICATIONS

This thesis contains an introductory review, followed by four research articles. In the introductory part, the articles are referred to by their roman numerals. Papers I, III, and IV are reprinted under the Creative Commons License. Paper II is reproduced with permission from the publisher.

I Svensson, J., Ström, J., Hansson, M., Lihavainen, H., and Kerminen, V.-M.: Observed metre scale horizontal variability of elemental carbon in surface snow. Environmental Research Letters, 8, 034012, doi:10.1088/1748-9326/8/3/034012, 2013.

II Svensson, J., Virkkula, A., Meinander, O., Kivekäs, N., Hannula, H.-R., Järvinen, O., Peltoniemi, J.I., Gritsevich, M., Heikkilä, A., Kontu, A., Neitola, K., Brus, D., Dagsson-Waldhauserova, P., Anttila, K., Vehkamäki, M., Hienola, A., de Leeuw, G., and Lihavainen, H.: Soot-doped natural snow and its albedo — results from field experiments. Boreal Environment Research, 21, 481-503, 2016.

III Meinander, O., Kontu, A., Virkkula, A., Arola, A., Backman, L., Dagsson-Waldhauserová, P., Järvinen, O., Manninen, T., Svensson, J., de Leeuw, G., and Leppäranta, M.: Brief communication: Light-absorbing impurities can reduce the density of melting snow, The Cryosphere, 8, 991-995, doi:10.5194/tc-8-991-2014, 2014.

IV Svensson, J., Ström, J., Kivekäs, N., Dkhar, N. B., Tayal, S., Sharma, V. P., Jutila, A., Backman, J., Virkkula, A., Ruppel, M., Hyvärinen, A., Kontu, A., Hannula, H.-R., Leppäranta, M., Hooda, R. K., Korhola, A., Asmi, E., and Lihavainen, H.: Contribution of dust and elemental carbon to the reduction of snow albedo in the Indian Himalaya and the Finnish Arctic, Atmospheric Measurements Techniques Discussion, in review, 2017.

AUTHORS' CONTRIBUTION TO THE PUBLICATIONS

I J. Svensson planned the study with J. Ström and M. Hansson. J. Svensson conducted the field measurements and the laboratory analysis. The results were interpreted by J. Svensson with supervision from J. Ström and M. Hansson. The manuscript was written by J. Svensson with contributions from other coauthors.

II A. Virkkula led the experiments with input from others. J. Svensson participated in the field experiments and analyzed data. J. Svensson prepared the manuscript with contributions from coauthors.

III O. Meinander formed the hypothesis presented in the paper, and wrote the manuscript. J. Svensson participated in collection of experimental data and analyzing field data, and contributed to the writing of the manuscript.

IV J. Svensson planned the study with J. Ström. Laboratory and field measurements were mainly conducted by J. Svensson. The results were interpreted by J. Svensson with supervision from J. Ström and input from others. The writing of the manuscript was led by J. Svensson with contributions from coauthors.

ABBREVIATIONS

BC	Black Carbon
eBC	Equivalent Black Carbon
BrC	Brown Carbon
CC	Carbonate Carbon
CCN	Cloud Condensation Nuclei
EC	Elemental Carbon
FMI ARC	Finnish Meteorological Institute
GAW	Global Atmosphere Watch
GrIS	Greenland Ice Sheet
IN	Ice nuclei
ISSW	Integrating Sphere Integrating Sandwich Spectrophotometer
LAHM	Light-Absorption Heating Method
LAI	Light-Absorbing Impurities
MAC	Mass Absorption Cross Section
MD	Mineral Dust
NIR	Near-Infra Red Region
OC	Organic Carbon
PSAP	Particle Soot Absorption Photometer
rBC	Refractory Black Carbon
SP2	Single Particle Soot Photometer
SoS	Soot on Snow
SZA	Solar Zenith Angle
TOM	Thermal-Optical Method

1. INTRODUCTION

Snow and ice are vital components in the Earth's spheres, interacting closely with the atmosphere, hydrosphere, and the biosphere. If snow is accumulated over a longer time span and the accumulation exceeds melting, the snow transforms into ice and a glacier begins to form. Glaciers have reshaped the modern world throughout the geological timescale with great mass fluctuations during glacial and interglacial periods. Today, most of the globe's frozen water is stored at the poles. Referred to as the third pole, the Hindu Kush-Himalayan and Tibetan plateau contains the most snow and ice outside of the Arctic and Antarctica. The water stored in glacier reservoirs worldwide accounts for about 70% of Earth's freshwater. With the vast amount of fresh water stored in the cryosphere, melting plays an integral part in regulating the water cycle. Melted glacier ice contributes to the current sea-level rise, while also some regions risk increased potential of natural hazards, such as landslides and glacial lake outburst floods.

Snow and glacier meltwater at mid latitudes and equatorial regions (e.g. South and North America and south and central Asia) provide a crucial fresh water source, necessary for agriculture, hydropower, and public health in many nations (e.g. Barnett et al., 2005; Immerzel et al., 2010; Kaser et al., 2010; Bolch et al., 2012). Meltwater delivers water during periods of drought. Water availability is, however, currently at risk due to climatic shifts. Additional climate changes could further strain water availability by changing the quantity and timing of melt. Such added stress, in regions with preexisting severe water scarcity, has the potential to create political instability and population migrations. Glaciers respond to climatic changes either through increased or decreased glacier volume, leading to an advancing or retreating glacier, respectively. But, since glacier changes are delayed by flow dynamics, there is a time lag between climatic changes and the response at the glacier terminus. In a warming climate the glacier mass balance typically will be negative, but the glacier runoff will increase and downstream areas have increased water availability. On the other hand, a continued shrinking and negative mass balance will affect glacier volume and eventually lead to less meltwater becoming available. The interactions between climate, cryosphere, and hydrosphere are complex and show dissimilar patterns world-wide. Thus, different areas face divergent fates. While some regions will experience a decrease in meltwater availability, others will confront the opposite, with increased water availability, depending on the stage of glacier change (Ragettli et al., 2016).

Fresh snow is a medium that reflects solar radiation more effectively than any other natural surface. This attribute is fundamental for Earth's climate. Covering immense land and sea areas in the northern hemisphere during parts of the year, snow and ice have an essential role in modulating the surface reflectance of incoming solar radiation. In this regard, the term albedo is of importance: defined as the amount of reflecting light divided by the incoming light. For visible light, this fraction of reflected light can exceed 90% (Warren, 1982). The rate and timing of snow and ice melt in the northern hemisphere have an important climatic role since uncovered ground or sea will have a significantly lower albedo than snow. This is most significant in the spring when snow extent is at its maximum and is also exposed to high solar insolation (e.g. Hall and Qu, 2006; Flanner et al., 2011). In the Arctic, the northern hemisphere snow albedo feedback has been observed and reported to be partially responsible for the Arctic amplification, where recent warming has been more pronounced than elsewhere globally (e.g. Déry and Brown, 2007; Serreze et al., 2007; AMAP, 2011).

While temperature increase is the main global driver of melting snow and glaciers, additional processes are also important (e.g. Barnett et al., 2005). Albedo depends on several factors for snow, but the primary agents are snow grain size and the presence of light-absorbing impurities (LAI)

(Warren and Wiscombe, 1980). A collective term for particulates that absorb light, LAI includes insoluble organic carbon (OC), mineral dust (MD), and the broadly defined black carbon (BC). Both OC and BC are derived from the incomplete combustion of carbonaceous matter, while MD originates from the crustal surface of the Earth. The tiny particles can be transported in the atmosphere over long distances before they are deposited onto snow and ice. Of the different impurities, BC is the material with the strongest climatic warming potential.

Once LAI decrease the snow albedo, several processes are set into motion which eventually cause a greater negative climatic effect. Snow with a lower albedo increases the amount of solar radiation absorbed, thus trapping even more heat in the snow. Warmer temperatures facilitates more rapid snow aging and a subsequent snow grain growth (Wiscombe and Warren, 1980). In larger grained snow BC has a more pronounced negative effect on the albedo compared to smaller grains (Warren and Wiscombe, 1980; Hadley and Kirchstetter, 2012; Dang et al., 2015). This positive feedback supplies itself with faster snow aging with higher temperatures, further reducing the albedo. Ultimately, this leads to an earlier onset of snowpack melt, uncovering the underlying surface with a significantly lower albedo, further hastening snow melt. During snow melt, LAI have a tendency to remain at the surface (e.g. Conway et al., 1996; Xu et al., 2012; Doherty et al., 2013), further concentrating their warming potential towards the surface of the snow, contributing to additional lowering of the albedo and speeding up melt (Flanner et al., 2007). Diminishing snow cover and increased thawing further influences the exchange between atmosphere and the ground, as well as the microbiology.

The overarching objective of this thesis is to investigate LAI in snow through field measurements and laboratory experiments in order to better understand and constrain the climatic impacts of LAI in snow. For this thesis, BC is emphasized, but other constituents are evaluated as well. Per unit mass, BC is the material with the largest potential impact on snow albedo, but anthropogenic and natural LAI must be studied jointly for a complete understanding. Further, the research conducted in this thesis focuses on the upper layers of the snowpack, where the albedo reduction from LAI is most applicable. While the negative effects of LAI on the cryosphere are clear, additional measurements and experiments are still needed. Measurements are sparse in some regions, and many of the process-level dynamics remain unresolved. An increased understanding will enable the scientific community to better constrain the effect of LAI on snow albedo and melt. For this purpose, different specific small-scale measurements and experiments have been conducted. The aim of this thesis is to examine the following questions:

1. What is the variability of black carbon in surface snow on spatial scales on the order of meters, and is there any difference in variability between polluted and clean sites?

Addressed in Paper I, this detail of small scale variability appeared to be overlooked in the pre-existing literature, thus Paper I aimed at contributing to this lack of knowledge.

2. What are the microphysical and radiative properties of an artificially perturbed snow surface using black carbon as light-absorbing impurity?

Dedicated experiments examining the relationship between LAI and the physical and radiative properties of snow are very limited. This is studied in Papers II and III.

3. How will light-absorbing impurities deposited during early spring affect the microphysical properties of the snowpack during the melting season?

Since BC has a tendency to remain in the top layers of the snowpack, it was hypothesized in Paper III that BC could influence snow melting properties.

4. Is it possible to deconvolute a laboratory liquid-based mixture of mineral dust and black carbon based on their respective optical properties, and how do the optical properties of the laboratory sample particles compare to ambient snow sample particles?

The optical properties of BC in snow have an effect on absorption, and it has been suggested that BC may be less absorbing than current estimates. This subject is investigated in Paper IV.

5. What is the relative contribution of mineral dust and black carbon to the light absorption of impurities at the high altitude glacier snow surface in Sunderdhunga valley, Himalayan India, and how does this contribution compare to the snowpack of the Finnish Arctic?

The current cryospheric changes occurring in the Himalaya are complex, and the role of the different LAI need to be scrutinized through measurements (e.g. Gertler et al., 2016). This is investigated in Paper IV.

2. BACKGROUND

2.1 Snow and interaction with radiation

Snowmelt is a product of the heat exchange between the snow and its surrounding environment. At the snow surface, temperature and the absorption of shortwave radiation mainly controls melt. Absorption depends on the incident radiation and the albedo. The albedo varies greatly with wavelength over the solar spectrum and depends on the physical properties of snow, such as grain size, depth, density, liquid water content, LAI, as well as cloud cover, and solar zenith angle (SZA) (Warren and Wiscombe, 1980). In the visible light spectra solar irradiance peak at the Earth's surface (most energy), and snow scatters visible light very efficiently and is basically non-absorptive. The deposition of nearly all aerosols on a snowpack therefore, apart from sulfate and sea-salt, will absorb sunlight producing a positive radiative forcing. In the near infra-red region (NIR), however, the effect of impurities is negligible as snow is dark and practically black, and the albedo is more sensitive to snow grain size (Warren, 1982). With time snow grains metamorphose, resulting in larger grains, enabling photons to travel deeper into the snowpack (Colbeck, 1982). With deeper penetration the photons are more likely to encounter a LAI, accounting for the greater negative effect LAI have in larger snow grains (Warren and Wiscombe, 1980; Hadley and Kirchstetter, 2012; Dang et al., 2015). The spectral properties of the snowpack have been used to deduce information about the surface properties using remote sensing techniques (e.g. Nolin and Dozier, 2000).

2.2 Light-absorbing impurities in snow

2.2.1 Carbonaceous aerosol

Aerosols refer to particles consisting of liquids or solids suspended in a gas. In the atmosphere, aerosols exists as a diverse combination of particles with different shapes, sizes, and chemical compositions. On an individual basis, a single particle can further consist of different constituents, with varying shapes and forms. Primary particles are directly emitted into the atmosphere, while secondary particles are formed in the atmosphere through gas to particle conversion. Emission sources for both types of aerosol can be either anthropogenic or natural, or a combination of both. An aerosol population usually encompass a wide size range, as aerosols exists in the range from a few nm up to tens of µm. The chemical and physical properties of an aerosol governs its interaction with surrounding media. Once in the atmosphere, the particles interact with light through reflection and absorption, contributing to what is known as the direct climate effect of aerosols. Aerosols also act as seeds to form cloud particles, so called cloud condensation nuclei (CCN) or ice nuclei (IN)

(Andreae and Rosenfeld, 2008). By altering the amount or properties of these nuclei particles, the microphysical properties of clouds may be perturbed. This may modify the optical properties or lifetime of clouds, known as the indirect climate effect of aerosols (Rosenfeld, 2000).

One aerosol species with a special relevance to the climatic impact of aerosols is the broadly defined BC. Black by nature, BC is the aerosol with the greatest absorption of light per unit mass. On a global scale, BC is estimated as second only to carbon dioxide as a climate warming agent (Jacobson, 2001; Hansen and Nazarenko, 2004; Ramanathan and Carmichael, 2008; Bond et al., 2013). It is produced during the combustion of carbon-based materials, largely fossil fuels and biomass burning. Depending on the specific carbon fuel used, temperature and pressure where the combustion occurs, an array of different particles produced can be classified as BC (Andreae and Gelencsér, 2006; Bond and Bergström, 2006; Bond et al., 2013; Petzold et al., 2013). Quantifying and characterizing the variety of BC particles is, therefore, not straightforward. The ideal analytical technique would determine several different properties of the particles simultaneously; currently this is not possible with a single instrument. As a consequence, different properties of BC are operationally defined dependent on measuring technique. Using the atmospheric research terminology, the different operational definitions proposed by Petzold et al. (2013) are: elemental carbon (EC) referring to methods that specifically measure carbon content of carbonaceous matter; refractory BC (rBC) is measured with incandescence methods; equivalent BC (eBC) when an optical absorption method has been used. Further, the term soot, refers to the process by which the particles are formed.

Black carbon does not only enhance climate warming, but is also detrimental to air quality and human health. Exposure to particles has been proven to have harmful health effects on humans (e.g. Janssen et al., 2011; Shindell et al., 2012). Determining emission sources and temporal variability of BC is therefore crucial from both a climate mitigation and air quality standpoint. Currently, global emissions estimates indicate an increasing trend, most probably due to expanding economies in Asian countries, whereas most other regions globally are decreasing their emissions (Bond et al., 2013). After a BC particle has been emitted it usually is hydrophobic (repels water molecules), but will rather quickly interact with other particles and condensable vapors to form a hydrophilic state (onto which water can adhere) (e.g. Jiao et al., 2014). Aerosol particulate mass has an average atmospheric residence time of one week in the lower troposphere, before it is scavenged, either through wet or dry deposition possibly far from its original emission source (Raes et al., 2000). Falling snowflakes are very efficient in scavenging aerosols (Paramonov et al., 2011). Microscopic studies of snowflakes have found thousands of aerosol particles inside or attached to snowflakes in Japan, and a large fraction of the aerosols were believed to be of carbonaceous origin (Magono et al., 1979). In instances when BC particles are mixed and rise to high altitudes their lifetime may be prolonged, which can transport them globally (Stohl, 2006). An aerosol co-emitted with BC during combustion and mainly biomass burning is OC, which may or may not absorb light. It is defined as brown carbon (BrC) when it absorbs light in the shorter wavelength ranges (Andreae and Gelencsér, 2006).

The mass absorption cross section (MAC) describes the efficiency by which aerosol particles absorb light. It is the light absorption coefficient divided by the mass concentration of the particles, usually expressed in $m^2\ g^{-1}$ (e.g. Seinfeld and Pandis, 2006; Moosmüller et al., 2009). This value depends mainly on the size of the particles. For airborne BC particles, MAC commonly has higher values for smaller particles (up to 100 nm), with a peaking MAC value in the range 100-200 nm, after which there is a sharp drop off in MAC with BC particle size (greater than 300 nm). This relates to smaller particles being volume absorbers of light, while the absorption of light not penetrating to the

core of the particle for larger particles. Measurements of MAC have been studied in detail in atmospheric aerosols (e.g. see review of Moosmüller et al., 2009), whereas the absorptivity of BC in snow is largely unresolved. Recently, an attempt was made to quantify LAI absorption in the seasonal snow of northern China, with the particulates collected on Nuclepore filters. This study made extensive use of the spectral response of different substances (Zhou et al., 2017). This method relied on filter techniques, which may cause different optical properties in ambient snow compared to filters conditions. Thus, the MAC inferred from filters may not accurately reflect values in snow conditions.

2.2.2 Mineral dust and other light-absorbing impurities

While BC is the particle with the most light-absorbing potential by mass in snow, other impurities existing in the snow also contribute to an albedo reduction and enhanced melt. The extent to which the impurity influence the snow is dependent of its absorptivity and concentration. Although the specific absorption by MD is less than BC, given enough MD concentration, the two can be of equal magnitude. Spectrally, the albedo reduction from BC is the greatest in the visible spectrum, while MD have a greater decrease in the Ultraviolet (UV) range (Warren and Wiscombe, 1980). Particles associated with MD originate from semi-arid regions where the soil is easily eroded and sparse vegetation can contain the particles from being suspended into the atmosphere. For global atmospheric aerosol mass and aerosol optical depth MD accounts for one of the most significant fractions (Tegen et al., 1997). It has the potential to be transported over great distances before depositing at Earth's surface and act as an important nutrient in the tropical rain forest, or perturb snow surfaces at the poles or even the Himalayas. Globally, there exists many areas that supply the atmosphere with MD, thus there are many different species of MD found world-wide in mixed variations in snowpacks and glaciers.

The significance of MD affecting snow albedo and melt has been investigated for some regions globally. It has been most extensively studied in the snowpacks of western United States, where it has been identified as the dominating LAI, shortening the duration of snow cover by weeks, as well as shifting the timing and intensity of runoff (e.g. Painter et al., 2007; Painter et al., 2010). In the Nepalese Himalaya, snow pit observations have shown that MD sometimes appear in such concentrations that it may be a greater than BC in disturbing snow albedo of Mera glacier (Kaspari et al., 2014). Likewise, glaciers of the European Alps have shown to be affected by MD originating from the Saharan desert (e.g. Lavanchy et al., 1999; Di Mauro et al., 2015; Gabbi et al., 2015). In the Arctic, Doherty et al. (2010) points out that 20-50 % of the light absorption by LAI in the seasonal snowpack is due to non-BC particles, although part of the fraction also includes BrC. Icelandic MD has been identified as a large source for the Arctic cryospehere (Meinander et al., 2016 and references therein), with the possibility of being distributed to the Greenland Ice Sheet (GrIS) (Groot Zwaaftink et al., 2016).

The term cryoconite is defined as granular sediment consisting of mineral and biological matter (Cook et al., 2015). Dark in appearance, cryoconite absorbs solar radiation, and is usually visible in centimeter deep meltwater holes on glacier and ice sheets world-wide. The role of microbiology and snow algae affecting snow albedo has recently been highlighted through measurements (e.g. Takeuchi et al., 2006; Lutz et al., 2014), and it appears that it can be significant for some regions. In the Arctic, for example, Lutz et al. (2016) estimated that 13% of the snow albedo decrease over one melt season was due to snow algae blooms.

2.2.3 Studies of light-absorbing impurities in snow

The effect of LAI on snow albedo was first outlined in detail through the modelling work of Warren and Wiscombe (1980). The work is based on Wiscombe and Warren (1980), where snow

reflectance modelling was over-estimated in the visible spectrum compared to observations. By incorporating small concentrations of LAI in the modelling, the model-measurement bias was eliminated (Warren and Wiscombe, 1980). More contemporary work, include the work of Hansen and Nazarenko (2004), which further raised awareness on BC in snow, by arguing that BC in snow may be responsible for 25% of the observed global warming. Additionally highlighted in Flanner et al. (2007), world-wide simulations showed the third pole as especially vulnerable, with an exceptionally high radiative forcing due to high deposition of LAI in snow.

To measure BC in snow with remote sensing techniques has been shown to be difficult, with surface properties introducing noise on the same magnitude as the actual BC reduction on albedo, making attribution of BC on albedo difficult (Warren, 2013). In-field measurements are therefore a crucial component of studying LAI effect on snow. The observational record of LAI in snow actually dates back prior to the remote sensing era. During the age of polar exploration, over a century ago, explorers such as Nordenskiöld and Nansen made the first recorded observations of impurities (Nordenskiöld actually first used the term cryoconite) in snow and ice on the GrIS. The modern day observational research on LAI in snow was outlined in the work of Clarke and Noone (1985). In their study concentrations of LAI in snow of the North American and European Arctic were found to be substantial enough to perturb the radiative balance of the snowpack. The spatial coverage was further updated in Doherty et al. (2010), extending with samples from many locations across the Arctic. This large spatial variability of BC in snow survey indicated that Greenland snow had the lowest concentrations, while the highest concentrations were in northeastern Siberia.

Spatial studies are good to obtain an overview representation of typical BC mixing ratios that can be observed on a large scale. However, it makes comparison difficult temporally since details concerning recent snowing and deposition of BC events are lost, which can have a large impact on the BC mixing ratios locally. For this purpose, ice cores are environmental archives that can provide deposition trends of LAI, even including pre-industrial times. In the Arctic the few existing records have indicated a peak in BC around 1910, coinciding with the period of industrialization (McConnell et al., 2007; Keegan et al., 2014; Ruppel et al., 2014). Since the 1970s, though, contrasting patterns for BC deposition has been observed (Ruppel et al., 2014). In the recent year of 2012, when the GrIS experienced exceptional wide-spread melt, BC has been suggested as significant contributor to this unusual melt (Keegan et al., 2014). The extent to which BC, as well as other LAI, is affecting the current albedo decay and melt increase of the GrIS is debated and complicated by many involved parameters and processes (e.g. Box et al., 2012; Dumont et al., 2014; Polachenski et al., 2015; Tedesco et al., 2016).

Long term records of LAI in snow from the Himalaya are also rather limited. The few records to-date have indicated contrasting patterns on the temporal progress of LAI in snow (e.g. Ming et al., 2008; Xu et al., 2009; Kaspari et al., 2011; Ginot et al., 2014; Jenkins et al., 2016). Snow pit studies investigating LAI, covering possibly a few annual deposition years, on the other hand, have provided spatial coverage of LAI in snow on the third pole (e.g. Xu et al., 2006, Ming et al., 2009; Kaspari et al., 2014; Jacobi et al., 2015; Yang et al., 2015; Li et al., 2016; Schmale et al., 2017). Most studies have shown an enriching of LAI at certain layers and that the LAI content decreases with altitude. The more recent work (e.g. studies of Li et al. and Schmale et al.) includes source apportionment, which provide indications on the origin sources for BC. Additionally, crude estimates on glacier melt from the LAI in the snow has been made (in Kaspari et al. and Schmale et al.). Contrasting patterns in the dominating impurity for albedo reduction has also been estimated, with MD in Nepal (Kaspari et al., 2014), while BC is more governing the snow albedo in central Asia and parts of Tibet (Schmale et al., 2017; Li et al., 2017).

Our current understanding of the effects of LAI on snow is largely based on models. Field experiments can help to better understand the interaction between LAI and snow. For BC on snow this has been done in outdoor conditions (Conway et al., 1996; Brandt et al., 2011), and in laboratory conditions (Hadley and Kirchstetter, 2012). In Conway et al. (1996) BC particles mixed in snow were spread over glacier snow surfaces in Washington State, U.S. followed by monitoring of the subsequent changes, with an emphasis on the movement of LAI within the snow with melt. In another outdoor experiment, artificial snow was produced with a snow-gun while BC was mixed with the freezing water (Brandt et al., 2011). With a highly contaminated snow this experiment confirmed radiative transfer calculations of the negative effect of BC on snow albedo. In a laboratory-based experiment Hadley and Kirchstetter (2012) showed that with increasing snow grain size the negative effect of BC on albedo was enhanced, compared to model predictions.

More detailed observations, on a process level in the snow, have suggested that BC particles have a tendency to be larger sized in snow compare to air (Schwarz et al., 2013). Information on the size of the particles is crucial for the absorptivity and the MAC of the BC. It also seems as if the larger airborne BC particles are more efficiently removed by precipitation (Schwarz et al., 2013; Mori et al., 2016). Once in the snow, the particle size appears to be further affected, with growth due to melt-freezing activities in the snow (Schwarz et al., 2013).

2.2.4 Measurement methods of light-absorbing impurities in snow

To measure all of the different LAI constituents in snow can be a complex task that requires more than one analytical technique. Ambient measurements are further challenged by the fact that the snow samples should be in a frozen state prior to analysis to avoid contamination (growth of biological material) or uncontrolled losses of particles. Maintaining the samples frozen is particularly challenging when snow samples are collected in remote environments, which they often are. Traditionally, as with airborne aerosols, LAI in snow have been collected on filters through filtering of melted samples. This approach eliminates the challenge of maintaining the sample frozen, since filtering can be done in field-like conditions. The filters can thereafter be analyzed optically, which has been done using an integrating sphere integrating sandwich spectrophotometer (ISSW) (Doherty et al., 2010; Grenfell et al., 2011). Using this measurement set-up an optical spectral absorption by impurities on the filter is obtained. It is not specific to the different constituents, instead it requires an interpretation of the spectral response. Another, less utilized method to analyze filters is the light absorption heating method (LAHM). It consists of exposing the particle-laden filters to visible light and measuring their absorptive response by monitoring the particle temperature increase, and has been used in the Andes of Peru (Schmitt et al., 2015). Another analysis technique for filters involves the thermal-optical method (TOM) (e.g. Forsström et al., 2009; Hagler et al., 2007; Cachier et al., 1989), and it is based on the different volatilization temperatures for different carbon species (OC and EC).

There is one existing non-filter based method, and it uses laser-induced incandescence of particles. The instrument used is the single particle soot photometer (SP2), (not explored in this thesis) which was originally made for atmospheric aerosols measurements. Particles contained in melted snow samples are needed to be aerosolized with a nebulizer before detection with the instrument, introducing uncertainties with this method (e.g. Schwarz et al., 2012). Nevertheless, the SP2 can provide actual size estimates of the BC particles, which is of great benefit for accurately calculating the absorption, and consequently, the radiative forcing in the snow.

Traditionally MD in snow has been measured by weighing filters before and after liquid samples has been filtered, applicable in regions where dust is believed to be the dominating LAI in

the snow occurring in higher concentrations (e.g. Aoki et al., 2006; Painter et al., 2012). Microscopy has also been used (Thevenon et al., 2009), as well as microparticle counters (Ginot et al., 2014) and mass spectrometry (Kaspari et al., 2014).

3. RESEARCH AREAS AND METHODOLOGY

3.1 Research areas

The geographical areas of research in this thesis consisted of different sites in Fennoscandia, resulting in the basis of the data for Paper I, III, IV, and the Indian Himalaya, constituting the data for Paper IV. Each site is characterized by different demographics and climatology, thus setting the stage for different investigations on LAI in snow. The Arctic and the Himalaya are two areas where LAI have been identified as having crucial roles in affecting the cryosphere (e.g. Hansen and Nazarenko, 2004; Flanner et al., 2007; Xu et al., 2009). Experimental campaigns in outdoor conditions on the seasonal snowpack at different locations in Finland are another part of this thesis, contributing with the measurements in Paper II and III.

3.1.1 Pallas and Sodankylä, Finland

The measurements originating from northern Finland are in the vicinity of the Pallas-Sodankylä Global Atmosphere Watch (GAW) station. Consisting of two main sites; the Pallas Atmosphere-Ecosystem Supersite and the Finnish Meteorological Institute's Arctic Research Centre in Sodankylä (FMI ARC). The former is located in northwestern Finland inside the Pallas-Yllästunturi national park, and the latter is situated 120 km southeast of Pallas. Both are north of the Arctic Circle. Snow samples from Pallas, used in Paper I and IV, were collected above the tree line in close proximity to the atmospheric measurement station that is positioned on top of the small fell, Sammaltunturi. Local emissions are minimal, but some influence from regional and long range transported particles exists. The snow at Pallas and Sodankylä is characterized as the northern boreal forest zone with a typical taiga snow type. Typically, snow cover lasts for about 200 days in the year (Oct-May), with maximum snow depth around 80-100 cm. At Sodankylä, weekly surface snow samples for LAI content have been collected since 2009. Parts of this data set has been used in this thesis. Sodankylä was also one of the experimental sites in the soot on snow (SoS) experiments, further explained in section 3.2. The snow at Sodankylä was investigated for Papers II, III, and IV.

3.1.2 Tyresta, Sweden

The Tyresta sampling site is located in Tyresta national park, about 25 km south-west of Stockholm, Sweden. With its close proximity to metropolitan Stockholm, and significant regional emissions, it served to compare the clean Arctic site with a polluted location in Paper I. Collection of snow samples was done from an open section of a mire in the spring of 2010, inside a larger forested zone with no known local emissions over a 5 km radius.

3.1.3 Sunderdhunga, India

The Sunderdhunga valley is located in the western Himalaya, in the state of Uttarakhand, India. Several glaciers are situated in the valley, but the chosen glaciers investigated in Paper IV were Bhanolti and Durga Kot glaciers (N 30° 12', E 79° 51'). Facing northeast they cover an elevation of 4400-5500 m a.s.l. The glaciers in the area contribute to the Ganges river basin, and there is no known local pollution. On a regional scale the closest towns are Bageshwar and Almora, 40 and 70 km southwards, with populations of 9 000 and 34 000, respectively. Otherwise the larger scale emission of particulates originate from the Indo-gangetic plain (IGP). Airborne measurements at Mukteshwar, 90

km southwards, have identified BC and dust to peak during the pre-monsoon season (Hyvärinen et al., 2011; Raatikainen et al., 2017).

3.2 Soot on snow experiments

Soot on snow (SoS) experiments were a series of outdoor experiments where soot was spread onto a natural snow surface and its effect on snow properties were monitored. The outcome from the experiments are presented Paper II and III. The first SoS experiment was on a farming field in southern Finland in Nurmijärvi, 30 km north of Helsinki. Soot was produced by burning wood and rubber pellets from used tires in a wood-burning stove. The smoke from combustion followed a pipe surrounded with snow for cooling into a rectangular experimental chamber placed on top of the snowpack where the particles were deposited. Next to the contaminated snow area a reference site was set-up. The following winters experiment was conducted at Jokioinen, in southern Finland about 100 km northwest of Helsinki. For this campaign a different approach to put impurities onto the snow surface was taken. Soot collected by chimney cleaners in Helsinki from wood and oil burning was deposited to the snow surface by a blowing system into a tent standing on top of the snow surface. Once the soot had been placed on the snow surface in circular spots of 4 m, unfavorable weather conditions seem to have masked the effect of soot on snow. With new snowfall and high winds our conclusion is that the top layer of the snow containing the soot was redistributed in the surrounding areas. Since this experiment did not provide quantitative data it will not be discussed in any further detail. The last experiment was made at the Sodankylä airfield, nearby the FMI ARC in Sodankylä, Arctic Finland. For this experiment several contaminant snow spots were made with different amounts of soot, and other LAI consisting of Icelandic volcanic ash and glaciogenic silt. The blowing system from 2012 was modified, otherwise contaminant spots were produced in the same way as in 2012.

3.3 Snow sample collection and filtering

From each location the collection of snow samples followed a similar working scheme. In field, snow layers were visually inspected, after which snow samples were usually collected with a stainless steel spatula into Nasco whirl-pak bags that had been tested not to contaminate samples. Subsequent melting and filtering of the sample is based largely upon the procedures from Forsström et al. (2009). Melting was conducted in a microwave oven, although a different melting procedure was utilized for part of the samples in Paper IV (details given below). The meltwater was filtered through glassware, with the impurities collected onto a micro quartz fiber filter (Munktell, 55 or 47 mm diameter, grade T 293). Filters were thereafter dried and stored in cold conditions before analysis.

Some site specific sampling were conducted and are worth mentioning in this context. For Paper I, snow samples were collected in different square grid-nets with 5 m or 2.5 m between each sampling point. This was done for surface snow samples (0-5 cm). For the experimental Papers II and III, snow surface samples were collected after the soot had been spread over the snow surface. In Paper IV, snow pits from two Indian glaciers were sampled in intervals, and the comparison snow from Finland contained surface snow samples from a seasonal snowpack. Due to the remoteness and high altitude of the samples obtained on glaciers in Paper IV, a different approach to the post-sampling handling was conducted. The location did not allow snow samples to remain frozen until melting and filtering in the laboratory. Therefore, the samples were melted over a camping stove in an enclosed glass container at the expedition base camp, after which they were filtered.

3.4 Light-absorbing impurities analysis

In this thesis the carbonaceous fraction of the LAI in snow was measured using TOM in Paper I, II, II, and IV. Thus, EC is the operationally defined constituent that has been measured. The Sunset laboratory OCEC-analyzer (Sunset Laboratory Inc. USA; Birch and Cary, 1996) was used. From the filter substrates a punch (usually either 1 or 1.5 cm^2) is taken and analyzed in two stages. During the first stage, in a helium atmosphere, the temperature is increased stepwise and OC and carbonate carbon (CC) is volatilized and detected by a flame ionization detector. In the second stepwise heating of the filter, oxygen is incorporated, and EC is measured. Since pyrolysis is likely to occur to some degree, a laser is used to measure the transmittance (and/or reflectance depending on instrument model) of the filter during analysis to account for potential charring. Different measurement protocols have been used, most notably NIOSH and EUSAAR_2 (Cavalli et al., 2010). The uncertainties with measuring LAI with TOM derive from inefficiency of the filters to capture all of the impurities, an uneven filter loading, as well as loss of particles to filtering equipment, most of which has been addressed to some degree in the different manuscripts and references within. Further, during filter analysis, filters containing a high MD loading can interfere with an accurate split point (Wang et al., 2012), and samples containing a high fraction of pyrolyzed OC can cause an artifact (Lim et al., 2014).

In Paper II electron microscopy study was added to investigate the LAI on a particle basis. For this purpose a sample of contaminated snow was melted onto a silicone disk. The remaining particles were thereafter observed with a Hitachi Hi-tech S-4800 field-emission electron microscope fitted with an Oxford Instruments Inca 350 energy-dispersive X-ray microanalysis system. Soot particles were identified as such by EDS measurements with both 5 kV and 20 kV acceleration voltages. The 5 kV measurements were used for detecting carbon and oxygen in the soot particles, and the 20 kV measurements were used to check for metals present in mineral dust, such as Na, Al, Ca, Fe.

To estimate the fraction of non-EC refractory impurities a custom built PSAP was added to the TOM analysis in Paper IV. Using this procedure has been reported previously by Lavanchy et al. (1999). However, as a different set of instruments were used here, a few laboratory procedure tests were conducted to verify the method, which are presented in Paper IV. The laboratory test included filters with different mixtures of LAI, which were generated in the laboratory by mixing different content of MD and BC with water followed by filtering the liquid mixture. Filter sets containing soot only, as well as dust only, and a mixture of the two were generated. The soot filters were made with two different kinds of soot: NIST and soot collected by chimney cleaners in Helsinki (same soot used in SoS experiments). For the mineral-containing samples two different minerals (Silicone carbide (SiC) and granite) were tested. The filters containing a mix of impurities were made with SiC and soot. The filters were analyzed according to the following procedure: from a dried filter a 1 cm^2 filter punch was measured with the PSAP, followed by OCEC-analysis. Thereafter it was again measured in the PSAP (while being compared to particle free filter). With this approach the change in transmission before and after burning off the carbonaceous impurities was obtained. This allowed fractions of MD to be estimated from the filters, since the light absorption from the carbonaceous constituents were made, leaving the remaining fraction to MD.

3.5 Albedo and physical properties of snow

In the SoS experiments measurements started immediately following soot deposition to the snow. In terms of albedo a set of pyranometers (manufactured by Kipp & Zonen) were employed for

this purpose by measuring the downwelling and upwelling irradiance at wavelengths 285 (or 310 depending on sensor) to 2800 nm. The albedo was the ratio between the two irradiances. Pyranometers measuring the upwelling irradiance were set 30 cm above the snow surface to measure throughout most of the day. The expanded standard uncertainty (2σ) was determined to ±2.8 % or ±6.1 %, depending on the sensor. Broadband albedo raw data had a time resolution of 1 min, but was reduced to a 1 h average (solar noon ±30 min) when presented in results and discussion section. This consistent approach also relived the measurement for any correction needed from shadows casted by the infrastructure for the pyranometers.

In the SoS experiments the snow physical properties observed consisted of thickness, density, hardness, grain size and shape. The measurements were made according to the International Classification for Seasonal Snow on the Ground (Fierz et al., 2009). In addition to these measurements, the last experiment had the additional snow grain size determined through macro-photography of each snow layer against a 1 mm grid.

4. RESULTS AND DISCUSSION

The main results of the research from the four papers encompassed in this thesis are presented and discussed in this chapter. The outline is as follows: section 4.1 presents the result from Paper I where the small scale variability of EC in surface snow was studied at both a clean and a polluted site; section 4.2 includes results from Paper II and III where a natural snowpack was experimentally contaminated with LAI. Specifically, section 4.2.1 focus on the albedo results, while section 4.2.2 highlights observations on the snow density made during the experiment. The results from Paper IV, where the contribution from different LAI constituents is investigated in snow from the Indian Himalaya and the Finnish Arctic, is presented in section 4.3; while the light absorption of EC from the laboratory and natural snow is assessed in section 4.4.

4.1 Small scale spatial variability of elemental carbon in surface snow

The investigated surface snow of Pallas displayed large varying EC concentrations on a horizontal meter scale of 2.5 and 5 m. The EC concentration had a range of 13 and 31 μg L^{-1} (25^{th} and 75^{th} percentile, respectively), and a standard deviation of 60% of the mean. At the comparison site near to urbanization, Tyresta, Sweden, with major regional emission sources (metropolitan of Stockholm), there was less relative spatial variability in the EC concentration in the snow surface with a range of 174 and 505 μg L^{-1} (25^{th} and 75^{th} percentile), and standard deviation of 20% of the mean. At Pallas, post-depositional processes in the snow are likely to be one main reason for the difference in small scale variability. The area sampled is above the tree line and snow drift is a common process with a key role in redistributing snow. Snowpacks near each other may, therefore, vertically represent very different EC depositional histories (i.e. EC concentration). This can create vertical gradients of EC in the snowpack, translating into horizontal spatial variability if wind erodes the surface and transports it to a different place. This may be especially evident when studying upper snow layers (top 5 cm of the snow in our case). Additionally, differences in the proximity to sources of BC can impact variability. Observations of airborne eBC have revealed polluted air masses originating from Central and Eastern Europe reaching Pallas, interceded with periods of clean air masses from the north, thus resulting in a heterogeneous temporal pattern of BC (Hyvärinen et al., 2011). On the contrary, at Tyresta, the sampling site receives higher EC concentrations in air on a more constant basis, which leads to greater dry and wet deposition on the snow surface, which may

mask any heterogeneity of EC in the snow. It is also very likely that the snowpack of Tyresta does not experience snowdrift to the same extent as Pallas.

Based on measurements conducted on the size of rBC particles in the snow, there are indications that there can be significant variability in the size distribution of rBC particles in snow. This seems to be due to the presence of exceptionally large rBC particles (Schwarz et al., 2012). The rate at which BC particles aggregate into larger particles is likely to be dependent on ambient meteorological conditions and the amount of BC deposited. Here, more frequent melt cycles and higher atmospheric BC concentrations are expected to lead to larger particles. As all the measurement techniques used today to characterize BC in snow have various size—dependent sampling efficiencies, this may introduce uncertainties. For the filter technique used in our study, there is an underestimation of small BC particles, which could attribute to the variability seen in the Pallas observations. The spatial variability of LAI in snow horizontally has been reported elsewhere (Forsström et al., 2013; Delaney et al., 2015; Lazarik et al., 2017), which further emphasizes the importance of collecting multiple samples from a measurement site to obtain a representative value.

4.2 Effect of soot on the physical properties of snow

4.2.1 Broadband albedo

After deposition of soot onto snow surfaces, the change in albedo was monitored and the temporal evolution could be compared between the different experiments. In 2011, the contaminated surface snow with an EC concentration of 20 000 µg L^{-1} had a general albedo decrease for the first week. Whereas in 2013, the snow spot containing the highest EC concentration (6 000 µg L^{-1}) first lowered the albedo when deposited on the snow, but during the subsequent two days the albedo recovered from approximately 0.4 to 0.65. A plausible reason for the different temporal evolutions of the albedo in the two experiment years could be that the decrease in 2011 was amplified by snow grain growth caused by the added soot particles. With larger grained snow, BC has an enlarged reduction of the albedo (Warren and Wiscombe, 1980; Hadley and Kirchstetter, 2012; Dang et al., 2015). The observed rising of albedo in 2013, on the other hand, could possibly be explained by soot particles sinking into the snow surface. This was on a millimeter to centimeter vertical scale and the particles sunk within minutes of deposition, during daytime when elevated solar radiation occurred. The daily maximum temperature was above 0°C during that time, while during the night it was -10°C (or below). The cold night temperatures possibly stopped further vertical motion of the absorbing particles within the snow. This process has further effects on the bidirectional reflectance factor (BRF), studied during the experiment (reported in detail in Peltoniemi et al., 2015). After the particles had depressed into the snow, the difference in surface reflectance between the contaminated snow and the clean snow is largest when measured at nadir, while it is smallest at larger zenith angles (where it is mostly pure snow one sees). This applies to higher impurity concentrations and implies that, for a ground observer, the albedo of contaminated snow could be overestimated, while the nadir angle (used for example by satellites) will underestimate the albedo (Peltoniemi et al., 2015).

After the first few days, the ensuing weeks showed similar patterns of albedo evolution for both experiments. New snowfall events covered the soot layer in the snow, increasing the albedo of the contaminated snow to a similar value as the albedo of the reference snow. The albedo of the snowpack containing soot particles was thereafter observed to decrease faster than the reference snow's albedo (e.g. during one event in 2011 the albedo was 0.83 and 0.82 after snow for the reference and contaminated snow, respectively. After two days the albedo was 0.66 and 0.36 for the reference

and contaminated snow, respectively). The buried soot layer therefore affected the new snow by increasing solar radiation and melting the new snow.

The observed albedo decrease during both the 2011 and 2013 experiments caused by increased BC concentration generally agrees with the previous laboratory study of Hadley and Kirchstetter (2012). In more detail, the SoS results show that the BC reduction on albedo in natural snow is not as pronounced as the laboratory conditions used in Hadley and Kirchstetter (2012). This is probably due to different light conditions used in each study (Hadley and Kirchstetter (2012) used a SZA of 0°). From the experimental data, a parameterization for albedo decrease with EC concentration was shown to be nearly identical to a similar function provided in Pedersen et al. (2015), which is based on extensive *in situ* measurements of spectral surface albedo and EC concentrations. The SoS data based parametrization was further used as input in the modelling work of Hienola et al. (2016), where the radiative impact of Nordic anthropogenic BC was estimated.

4.2.2 Snow density

Snow density is generally understood to increase with time and especially during melt season. Counterintuitive to this notion, the snow density was observed to decrease during snowmelt in the SoS experiment. Based on the concurrent measured LAI content in the snow, it appeared to be linked to the LAI concentration, as higher LAI content was associated with lower snow densities. The same relationship between increasing LAI content and decreasing density was not observed in non-melting snow. During the snowmelt period, a rain event on snow occurred. However, the same conditions where maintained with lower densities for increasing LAI concentration. Based on this observation, it was hypothesized that LAI decrease the water retention capacity of melting snow. Additional, independent, small scale experiments on the meltwater release confirmed that LAI contaminated snow discharge water faster than clean snow. Plausible processes explanations for the decrease in snow density could be that LAI:

1. Increase melting or evaporation or both of the snow surrounding the impurities, allowing air pockets around the impurities to be formed, leading to less dense snow
2. Reduce the adhesion of water to the snow grains. The decrease of liquid water holding capacity will result in decreased snow density.
3. Increase snow metamorphism, and consequently increasing grain size, which has a lower water retention capacity, creating a reduced density.

In natural snow melting conditions at Sodankylä, following the experiments, analogous snow density decrease with increasing LAI has been observed to occur (Meinander et al., 2016). Recently, Skiles and Painter (2017), reported from measurements in the seasonal snowpack of Colorado, U.S., a similar lowering of surface snow density in the presence of a heavy dust loading. Before robust conclusions can be made on this density effect, additional measurements are needed.

4.3 Contribution of different light-absorbing impurities in snow

The investigated glacier area in the Indian Himalaya, Sunderdhunga (an area previously unexplored with respect to LAI in snow) displayed higher EC concentrations than previous measurements of EC from inside the Chinese boarder to the northeast. Although direct comparisons are difficult due to different sets of measurement techniques and surrounding geography, Sunderdhunga may have a greater amount of LAI in the snow, due to its lower altitude and location on the southern flank of the Himalaya.

The fraction of MD responsible for the light absorption on the filter was very high for the Indian originating filters. The majority of surface snow samples had MD contributions exceeding

56%, thereby indicating an abundance of dust in the snow on the two sampled glaciers. Hence, MD probably plays an important role in reducing the snow albedo on glaciers in this area of the Himalaya. On the contrary, the two investigated sites in the Finnish Arctic displayed a much smaller fraction of MD for the LAI in the snow. Most MD fractions were smaller than 20%, which is on the same scale as estimates reported from other Arctic locations (Doherty et al., 2010). In this part of the Arctic, MD is likely to affect the snow radiative balance to a much lesser extent than the Himalayan samples. Other studies have highlighted the importance and abundance of MD in other regions of the Himalaya, where in some cases dust has been measured at such high concentrations that it may be exceeding BC in the snow albedo reduction as the most dominant particulate (Kaspari et al., 2014; Qu et al., 2014). Contrasting observations have been made in other third pole regions however, with BC being the dominant light absorber in the snow (Ming et al., 2016; Li et al. 2017; Schmale et al., 2017; Zhang et al., 2017). A broader picture of MD versus BC in reducing snow albedo is still needed in high mountain Asia.

4.4 Optical properties of elemental carbon in snow

The measurement obtained MAC EC for the ambient samples from Arctic Finland and Himalayan India is roughly half of the MAC for laboratory EC particles treated with the same liquid-filtering procedure. This indicates a less absorbing efficiency for the same amount of mass EC for the snow originating particles. Hypothetical speculations for the difference include a possible difference in the size of EC particles, with the laboratory filters containing a higher amount of smaller sized EC particles compared to the ambient samples. A higher frequency of smaller particles will result in a greater MAC. Another hypothesis to explain the difference in MAC is that a higher content of OC was measured in the snow originating EC particles. Elevated OC content, combined with the EC particles being further embedded in the filter medium due to liquid-filtering, could have the effect of masking the EC particles, making them less efficient absorbers in this particular matrix. This is inconsistent with results from air samples (e.g. Jacobson, 2001; Cappa et al., 2012; Bond et al., 2013). In a snow albedo context, EC particles with less absorptive efficiency have implications on the snow albedo reduction, which has also been suggested in Schwarz et al. (2013). Based on their observations of the size of rBC particles in snow, Schwarz et al. calculate that the BC absorption in snow is overestimated by 40%.

5. CONCLUSIONS

In this thesis a set of different measurements and experiments were conducted to increase our understanding of LAI interaction with snow and its role in the climate system. In conformance with the aforementioned scientific questions, the main conclusions of this thesis are:

1. The BC concentration in surface snow presented large horizontal variability, significant down to the meter scale. It was clear that the pristine Finnish Arctic presented a relatively larger variability compared to the more polluted site near Stockholm. Differences in post-depositional processes of BC and distance to emission sources are pointed out as main reasons for the differences in variability.
2. Following BC doping of a snow surface, the albedo of a natural snowpack is reduced. With higher BC concentrations, on the order of >1 000 μg L^{-1}, the decrease in albedo and change in snow properties can be attributed to the added LAI. At smaller concentrations, the BC effect may be masked by other influences (external such a meteorological conditions or other

micro-processes in the snowpack). The importance of observation at very high temporal frequency cannot be overstated.

3. The density of surface snow decreases with increasing LAI concentration during melt. No such relationship was observed in non-melting conditions. The LAI likely reduce the water retention capacity of the melting snow, thereby decreasing the density.
4. Through optical measurements of particle-laden filters combined with mass estimates, the respective fractions of MD and BC can be assessed, when combining thermal-optical and strictly optical methods. The light absorption of BC particles originating from snow has half of the light absorbing efficiency of laboratory based BC particles. In order to more accurately estimate the BC forcing in snow, the site-specific optical properties of BC should be considered.
5. Deposited LAI to the Himalayan glacier surface at an elevation of 5000 m exists in highly concentrated annual layers that affect the radiative balance of the glacier surface. At a higher elevation the contribution of LAI is unknown. The fraction of MD absorbing light can sometimes exceed that of BC, suggesting that the snow radiative balance in the sampled area may at times be more affected by MD than BC.

6. FUTURE OUTLOOK

The conducted measurements and experiments in this thesis have underlined areas where further research is needed. One suggested topic is the process of LAI deposition from the atmosphere to the snow surface. Measurements of deposition and the removal of LAI from the atmosphere to the snow would allow new perspectives on this process, and would improve our understanding of LAI interaction with the snow surface. Once the particulates are at the snow surface, it is vital to investigate LAI migration through the snowpack. Traditionally, LAI has been observed to remain at the surface during melt. However, this process appears required to be re-evaluated (Lazarcik et al., 2017). Additional post-depositional processes in the snow, such as BC growth in size by agglomeration is another subject that would benefit additional studies. The light absorption of BC in snow (as well as other LAI), a parameter very influential for the reduction in albedo, needs to be further addressed. This will enable more accurate estimates on the LAI reduction of snow albedo. Ideally the MAC would be measured while the LAI particles are in the snowpack, since the light condition will change in other measuring environments (e.g. particles collected on filter). Although the task to measure in the snow is challenging, as ideally non-intrusive measurement techniques would be desirable to investigate LAI and snow interaction on a micro scale.

A geographical area, where LAI and the different constituents are affecting the cryosphere, needing further attention is the Himalaya and the other parts of the third pole. This vast region has divergent patterns for both glacier change and LAI deposition, and in order to assess the role of LAI affecting the area, supplementary research is necessary. Likewise, the Andes of South America has not been explored in terms of LAI affecting the ongoing negative cryospheric changes.

In the future the relative contribution between natural and anthropogenic LAI in snow is not very clear. Climate change may increase natural deposition of MD and biomass BC from forest fires, whereas political incentives may reduce the BC from fossil fuels. Any scenario requires detailed knowledge about the processes acting on the snow at the grain size level in order to understand the big picture about the cryosphere and the hydrological cycle. More process research is needed in order to provide climate models with the best available understanding. Our prediction skills about our future environment will never be better than the knowledge we feed into the models.

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